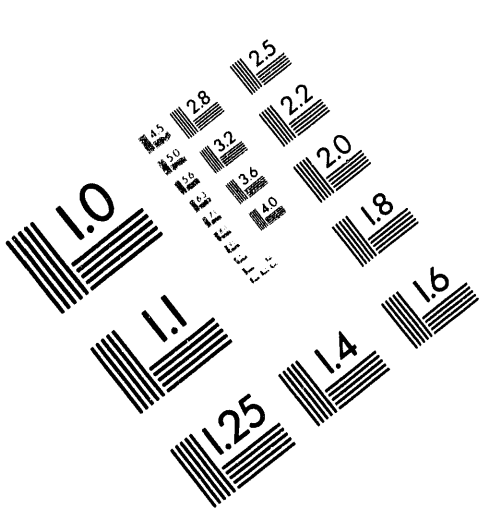
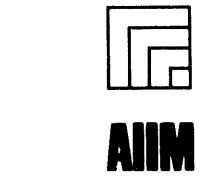
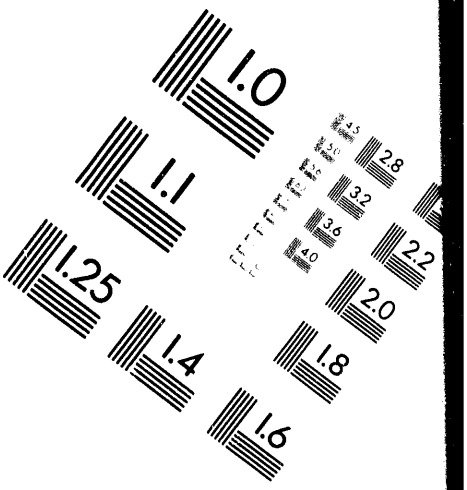
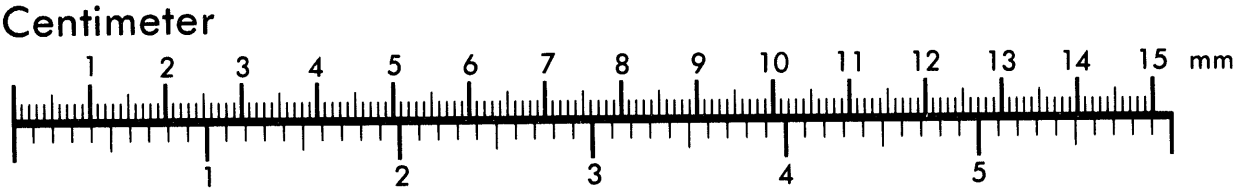
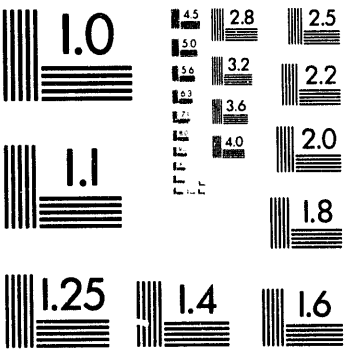


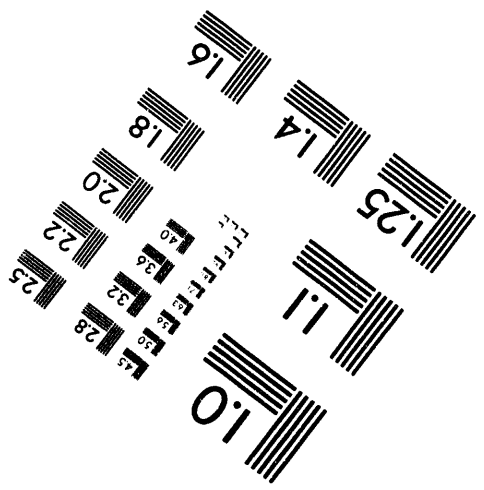


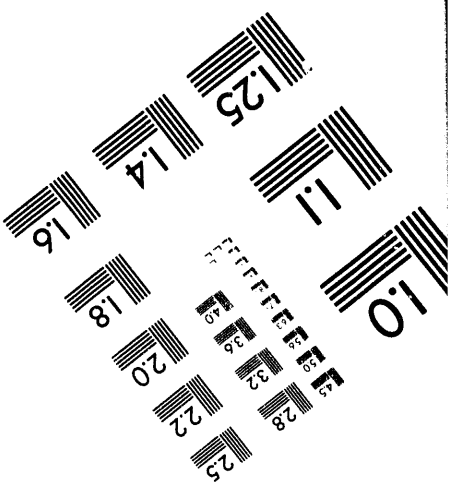
Association for Information and Image Management
1100 Wayne Avenue, Suite 1100
Silver Spring, Maryland 20910
301/587-8202











MANUFACTURED TO AIIM STANDARDS
BY APPLIED IMAGE, INC.

1 of 1

Conf-940749--59

LA-UR- 94-2375

Title: COMPARISON OF THREE GAMMA RAY ISOTOPIC DETERMINATION CODES: FRAM, MGA, and TRIFID

Author(s): Teresa L. Cremers
Jack E. Malcom
Charles A. Bonner

Submitted to: The Institute of Nuclear Materials Management (INMM)
July 17-20, 1994
Naples, Florida

Los Alamos
NATIONAL LABORATORY

Form No. 836 R5
ST 2629 10/91

COMPARISON OF THREE GAMMA RAY ISOTOPIC DETERMINATION CODES: FRAM, MGA, AND TRIFID

Teresa L. Cremers, Jack E. Malcom, and Charles A. Bonner, Los Alamos National Laboratory, Los Alamos, New Mexico, 87545 USA

The determination of the isotopic distribution of plutonium and the americium concentration is required for the assay of nuclear material by calorimetry or neutron coincidence counting. The isotopic information is used in calorimetric assay to compute the effective specific power from the measured isotopic fractions and the known specific power of each isotope. The effective specific power is combined with the heat measurement to obtain the mass of plutonium in the assayed nuclear material. The response of neutron coincidence counters is determined by the ^{240}Pu isotopic fraction with contributions from the other even plutonium isotopes. The effect of the ^{240}Pu isotopic fraction and the other neutron contributing isotopes are combined as ^{240}Pu effective. This is used to calculate the mass of nuclear material from the neutron counting data in a manner analogous to the effective specific power in calorimetry[1]. Comparisons of the precision and accuracy of calorimetric assay and neutron coincidence counting often focus only on the precision and accuracy of the heat measurement (calorimetry) compared to the precision and accuracy of the neutron coincidence counting statistics[2]. The major source of uncertainty for both calorimetric assay and neutron coincidence counting often lies in the determination of the plutonium isotopic distribution as determined by gamma ray spectroscopy. Thus, the selection of the appropriate isotopic distribution code is of paramount importance to good calorimetric assay and neutron coincidence counting. Three gamma ray isotopic distribution codes, FRAM[3] (Version 2.1), MGA[4,5] (Version 1.03), and TRIFID[6] (Version 5.89.1), have been compared at the Los Alamos Plutonium Facility under carefully controlled conditions of similar count rates, count times, and ^{240}Pu isotopic fraction.

EXPERIMENTAL

The three codes differ in the energy ranges analyzed, the number of detectors used, and peak area and isotopic ratio calculational methods. The data collection hardware used with all three codes was set up and operated according to the code developers specification. FRAM uses a single planar detector to obtain spectral data in the 120-420 keV range. TRIFID analyzes spectral data from 120-450 keV obtained with a planar detector and 630-725 keV obtained with a coaxial detector. MGA analyzes spectra data from 60-300 keV obtained with a planar detector and above 300 keV with a coaxial detector. The amplifier gains and shaping constants for the three systems are described in Table I, below. Pile up rejection circuitry was used in all cases. All spectra were collected in "autocycle" mode, that is, one spectrum was collected after another automatically without sample replacement. In all cases, the ^{242}Pu isotopic fraction were obtained from the mass spectroscopy data and not calculated from the codes. Samples were rotated and translated vertically during data collection. Count rates were controlled by moving the detectors closer or farther from the samples as required. Data were collected and the spectra were analyzed without assistance of the code developers. This allowed the robustness and "friendliness" of the codes to be tested under actual operating conditions.

Table I. Amplifier gains and shaping constants used for data collection.

Code	Detector	Amplifier Gain (keV/channel)	Shaping Constant (μsec)
FRAM	planar	0.1025	1
MGA	planar	0.075	2
	coaxial	0.250	2
TRIFID	planar	0.115	2
	coaxial	0.258	1

Spectral data were obtained for six standard items. Unfortunately, the data set is not complete because the Los Alamos Plutonium Facility was closed before all of the measurements could be taken. Mass spectral data were available to provide reference values for the plutonium isotopic fractions. The ^{240}Pu isotopic fraction of these six items varied from six to forty percent. The items 20LOAM and 20HIAM were from the same blended parent batch of plutonium oxide but the americium had been removed by chemical processing from the item 20LOAM. Thus, these two items have identical plutonium isotopic fractions, but very different americium concentrations. The items 40LOAM and 40HIAM were treated similarly and are a low and high americium pair with identical plutonium isotopic fractions. The nominal plutonium isotopic fractions and americium concentrations of the six standards are presented in Table II.

Table II. Plutonium isotopic fractions and americium concentration of isotopic standards as of January 1, 1994.

	^{238}Pu (wt%)	^{239}Pu (wt%)	^{240}Pu (wt%)	^{241}Pu (wt%)	^{242}Pu (wt%)	^{241}Am (μg/g Pu)	P_{eff} (mwatt/g)	$^{240}Pu_{eff}$ (fraction)	g Pu
CALEX	0.009	93.92	5.86	0.184	0.029	1917	2.503	0.0593	398
STD40	0.062	87.32	11.79	0.631	0.201	6285	3.612	0.1228	869
20LOAM	0.423	74.91	20.81	2.159	1.701	831	5.487	0.2473	422
20HIAM	0.423	74.91	20.81	2.159	1.701	43435	10.352	0.2473	653
40LOAM	0.381	53.74	37.83	3.827	4.225	1340	6.164	0.4589	315
40HIAM	0.381	53.74	37.83	3.827	4.225	84905	15.708	0.4589	547

RESULTS AND CONCLUSIONS

The spectral data were analyzed and the values for P_{eff} and $^{240}Pu_{eff}$ were compared to the standard values calculated from the mass spectroscopy data for the plutonium isotopic fractions. The

uncertainties calculated are the standard deviations of the mean ratios ($\sigma_{mean} = \sigma_{single\ measurement}/\sqrt{N}$, where N is the number of measurements). The standard deviations of single measurements are also given in the data tables.

The data presented in Table III show that all three codes are essentially unbiased for P_{eff} and $^{240}Pu_{eff}$ for measurements of low burnup (6% ^{240}Pu) material. The single measurement precision obtained with MGA is slightly better for both P_{eff} and $^{240}Pu_{eff}$. This is as expected because MGA analyzes data from the intense 100 keV region of the spectrum and is able to obtain good counting statistics. Differences in the precision for $^{240}Pu_{eff}$ for MGA and TRIFID are not significant.

Table III. Measurement of standard CALEX . Count rate = 5000 Hz on FRAM, TRIFID, and MGA planar detectors. Count rate on TRIFID coaxial detector = 11,000 Hz. Count rate on MGA coaxial detector = 5000 Hz. True count time = 7200 seconds.

		Specific Power		^{240}Pu Effective Fraction	
CODE	Number of Meas.	Ratio: Meas/Cert	%RSD Single Measurement	Ratio: Meas/Cert	%RSD Single Measurement
FRAM	10	1.000 ± 0.0012	0.38	1.005 ± 0.0073	2.3
MGA	10	0.997 ± 0.0007	0.23	0.993 ± 0.0028	0.9
TRIFID	10	1.002 ± 0.0010	0.31	1.004 ± 0.0032	1.0

The data for the specific power for the 12% ^{240}Pu isotopic fraction standard, STD40, show that the precision and bias of FRAM and MGA are very similar. TRIFID shows a small bias, approximately two percent. For the $^{240}Pu_{eff}$ measurements, MGA delivers the best precision and bias. The precision and bias of FRAM and TRIFID for $^{240}Pu_{eff}$ are the same. The data for STD40, 12% ^{240}Pu, are presented in Table IV.

Table IV. Measurement of standard STD40. Count rate = 30000 Hz on FRAM, TRIFID, and MGA planar detectors. Count rate on TRIFID and MGA coaxial detectors = 30000 Hz. True count time = 3600 seconds.

		Specific Power		^{240}Pu Effective Fraction	
CODE	Number of Meas.	Ratio: Meas/Cert	%RSD Single Measurement	Ratio: Meas/Cert	%RSD Single Measurement
FRAM	15	1.0029 ± 0.0006	0.23	1.007 ± 0.0028	1.1
MGA	15	0.9964 ± 0.0004	0.16	0.998 ± 0.0010	0.4
TRIFID	15	1.0200 ± 0.0005	0.19	1.014 ± 0.0021	0.8

The data for both 20LOAM and 20HIAM for specific power show that FRAM gives slightly better precision and accuracy than TRIFID (there were no data available for MGA). For

measurement of ^{240}Pu effective fraction both codes deliver results that are approximately the same. The data for 20LOAM and 20HIAM are presented in Tables V and VI.

Table V. Measurement of standard 20LOAM. Count rate = 20000 Hz on FRAM and TRIFID planar detectors. Count rate on TRIFID coaxial detector = 20,000 Hz. True count time = 3600 seconds.

		Specific Power		^{240}Pu Effective Fraction	
CODE	Number of Meas.	Ratio: Meas/Cert	%RSD Single Measurement	Ratio: Meas/Cert	%RSD Single Measurement
FRAM	15	1.0042 ± 0.0009	0.36	1.0002 ± 0.0026	1.03
TRIFID	15	1.0320 ± 0.0012	0.49	1.0082 ± 0.0014	0.54

Table VI. Measurement of standard 20HIAM. Count rate = 30000 Hz on FRAM and TRIFID planar detectors. Count rate on TRIFID coaxial detector = 30000 Hz. True count time = 3600 seconds.

		Specific Power		^{240}Pu Effective Fraction	
CODE	Number of Meas.	Ratio: Meas/Cert	%RSD Single Measurement	Ratio: Meas/Cert	%RSD Single Measurement
FRAM	15	1.014 ± 0.0005	0.19	0.985 ± 0.003	1.02
TRIFID	15	1.040 ± 0.0008	0.31	1.004 ± 0.003	1.27

The standard 40HIAM (Table III, 38% ^{240}Pu) presented an unexpected challenge. Although putatively pure plutonium oxide, this material proved to be contain approximately 0.2 percent ^{237}Np as an impurity. All of the codes detected the neptunium (easily confirmed by examination of a gamma ray spectrum of the material), but the precisions and accuracies presented in Table VII are worse than those for the other standards. FRAM gives the most precise and accurate measurements for this material. Both MGA and TRIFID are significantly biased.

Table VII. Measurement of standard 40LOAM. Count rate = 30000 Hz on FRAM, TRIFID, and MGA planar detectors. Count rate on TRIFID and MGA coaxial detectors = 30,000 Hz. True count time = 3600 seconds.

		Specific Power		^{240}Pu Effective Fraction	
CODE	Number of Meas.	Ratio: Meas/Cert	%RSD Single Measurement	Ratio: Meas/Cert	%RSD Single Measurement
FRAM	15	1.0061 ± 0.0005	0.18	0.994 ± 0.0013	0.51

MGA	15	0.9826 ± 0.0011	0.44	0.978 ± 0.0009	0.34
TRIFID	15	1.0400 ± 0.0009	0.36	1.023 ± 0.0019	0.72

The standard 40HIAM was particularly difficult because of the high americium (8.5%) concentration. Visual examination of the gamma spectrum revealed an intense continuum, probably arising from the high neutron flux from the sample, across the spectrum. Of the two codes tested with this material, FRAM and TRIFID, FRAM gave better performance for both specific power and ^{240}Pu effective fraction. The data are presented in Table VIII.

Table VIII. Measurement of standard 40HIAM. Count rate = 20000 Hz on FRAM and TRIFID planar detectors. Count rate on TRIFID coaxial detector = 20000 Hz. True count time = 3600 seconds.

		Specific Power		^{240}Pu Effective Fraction	
CODE	Number of Meas.	Ratio: Meas/Cert	%RSD Single Measurement	Ratio: Meas/Cert	%RSD Single Measurement
FRAM	15	0.987 ± 0.0066	2.51	1.0001 ± 0.016	4.49
TRIFID	15	1.107 ± 0.0213	9.14	0.9873 ± 0.032	12.32

The samples 40LOAM and 40HIAM also presented an opportunity to investigate the effects of count rates on the precision and accuracy of the measurement of specific power and ^{240}Pu effective fraction. For the low americium sample, 40LOAM, the precision of both specific power and ^{240}Pu effective fraction varied as a function of counting statistics, as expected for the count rates. For the high americium sample, 40HIAM, the precision of the specific power and ^{240}Pu effective fraction did not change very much as the count rate was changed. This is effect probably caused by the high continuum observed in 40HIAM spectra and the concomitant corruption of the spectrum and counting statistics. These data are presented in Tables IX and X for the FRAM code. The data for MGA and TRIFID are not complete and are not given here, but the available unpublished data suggests that a similar effect would be observed for these codes, too. The data presented in Table X suggest that for materials with marginal spectra, an increase in count rate or count time will not improve the analysis.

Table IX. Precision and accuracy of specific power and ^{240}Pu effective fraction as a function of count rate for FRAM code analysis of 40LOAM spectra. True count time = 3600 seconds.

		Specific Power		^{240}Pu Effective Fraction	
Rate (kHz)	Number of Meas.	Ratio: Meas/Cert	%RSD Single Measurement	Ratio: Meas/Cert	%RSD Single Measurement
10	15	1.0041 ± 0.0010	0.37	1.0019 ± 0.0023	0.90

20	15	1.0024 ± 0.0008	0.31	1.0023 ± 0.0014	0.53
30	15	1.0061 ± 0.0005	0.19	1.0046 ± 0.0013	0.51
40	15	1.0053 ± 0.0005	0.20	1.0029 ± 0.0010	0.37
50	15	1.0050 ± 0.0004	0.15	0.9996 ± 0.0008	0.33

Table X. Precision and accuracy of specific power and ^{240}Pu effective fraction as a function of count rate for FRAM code analysis of 40HIAM. True count time = 3600 seconds.

		Specific Power		^{240}Pu Effective Fraction	
Rate (kHz)	Number of Meas.	Ratio: Meas/Cert	%RSD Single Measurement	Ratio: Meas/Cert	%RSD Single Measurement
10	15	0.9942 ± 0.0057	2.21	0.9993 ± 0.0097	3.77
20	15	0.9873 ± 0.0066	2.51	1.0001 ± 0.0116	4.49
30	15	0.9986 ± 0.0051	1.97	0.9749 ± 0.0105	3.97
40	15	0.9939 ± 0.0067	2.58	0.9784 ± 0.0104	3.93

The comparison data indicate that the selection of the isotopic code can be optimized with knowledge of the ^{240}Pu isotopic fraction and the americium concentration of the nuclear material to be analyzed. The code FRAM gives excellent results for a wide variety of materials and would be a good choice for a facility where many types of materials are assayed. Pure metals and oxides of low burnup plutonium (~ 6% ^{240}Pu) are best analyzed with MGA. The ^{240}Pu effective fraction of intermediate burnup material (~ 20%^{240}Pu) can be determined with good precision and bias by TRIFID. Finally, for materials with poor spectra, increasing the count rate or count time may not lead to improved assay precision.

REFERENCES

[1] REILLY, T.D., ENSSLIN, N., and SMITH, H.A., Jr., Eds., "Passive Nondestructive Assay of Nuclear Materials", NUREG/CR-5550 (U.S. Nuclear Regulatory Commission, Washington DC, 1991).

[2] SAMPSON, T.E., "Precision of Gamma-Ray Measurements of the Effective Specific Power of and Effective ^{240}Pu Fraction of Plutonium", Los Alamos National Laboratory report LA-12328-MS (May 1992).

[3] SAMPSON, T.E., NELSON, G.W., and KELLEY, T.A., "FRAM: A Versatile Code for Analyzing the Isotopic Composition of Plutonium from Gamma-Ray Pulse Height Spectra", Los Alamos National Laboratory report LA-11720-MS (December 1989).

[4] GUNNINK, R., "MGA: A Gamma-Ray Spectrum Analysis Code for Determining Plutonium Isotopic Abundances", Lawrence Livermore National Laboratory report UCLR-LR-103220, Vol. 1 (ISPO No. 317, Task No. A.161) (April 1990).

[5] KEONIG, Z.M., BUCKLEY, W.M., RUHTER, W.D., GUNNINK, R., and CAMP, D.C., "Actinide Isotopic Analysis System for Los Alamos Plutonium-Handling Facility, Inst. Nucl. Mater. Mange. **XX**, 470-473 (1991).

[6] FLEISSNER, J.G., CORESSEL, T.W., FREIER, D.A., and MACKLIN, L.L., "TRIFID, A Second Generation Plutonium Isotopic Analysis System", Inst. Nucl. Mater. Manage. **XVIII**, 814-820 (1989).

DATE

FILMED

9/8/94

END